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レーザーで遊ぶ非線形振動

Nonlinear Oscillation Induced by Laser

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Living organisms on the earth maintain their lives under thermodynamically open condition by accepting the energy supply from the sun. Thus, studies on the generation of spatio-temporal structure under light illumination would be of scientific value not only in basic physics but also in biological sciences. As typical characteristics generated under thermodynamically open condition, breaking of the time-translational symmetry, or appearance of temporal rhythm, would be important in relation to the dynamic aspect of life.[1,2] There are rich varieties of examples on symmetry breaking of time translation in living matter; beating heart, nervous firing, circadian rhythm, cell-cycle, etc., where the periodicity falls, on the order of ms ~ day.[3] As for non-biological systems, self-pulsing of laser is the representative oscillatory phenomenon generated under far-from-equilibrium conditions, where the periodicity is rather short, being on the order of ns ~ μs.[4-6] In the present paper, we would like to report a novel rhythmic phenomenon on the periodic growth and burst of the cluster of submicrometer-sized polystyrene beads in aqueous solution under the illumination of focused Nd:YAG IR laser beam.

As has been indicated by Ashkin, an object can be trapped with focused laser, laser trapping.[7] In general, force generated with focused light is represented as the summation of light gradient and scattering terms,[8,9]

$$\mathbf{F}(\mathbf{r}) = \alpha\nabla|\mathbf{E}(\mathbf{r})|^2 + \beta\mathbf{E}(\mathbf{r})\times\mathbf{H}(\mathbf{r}). \qquad (1)$$

Where α and β are constants, as the function of dielectricity, refractive index of medium, and velocity of light. The first term describes an attractive potential around the focus, where the attractive force decreases with the sharpening of angle of the optical cone (see Fig. 1a). On the contrary, as in the last term in eq. (1), the scattering force exerting on an object increases with the decrease of the cone angle. It is, thus, expected that instability on optical trapping is induced with the decrease of the cone angle.[10] Bearing such effect on mind, we have carried out the experiment on laser trapping of submicrometer-sized plastic beads bearing negative

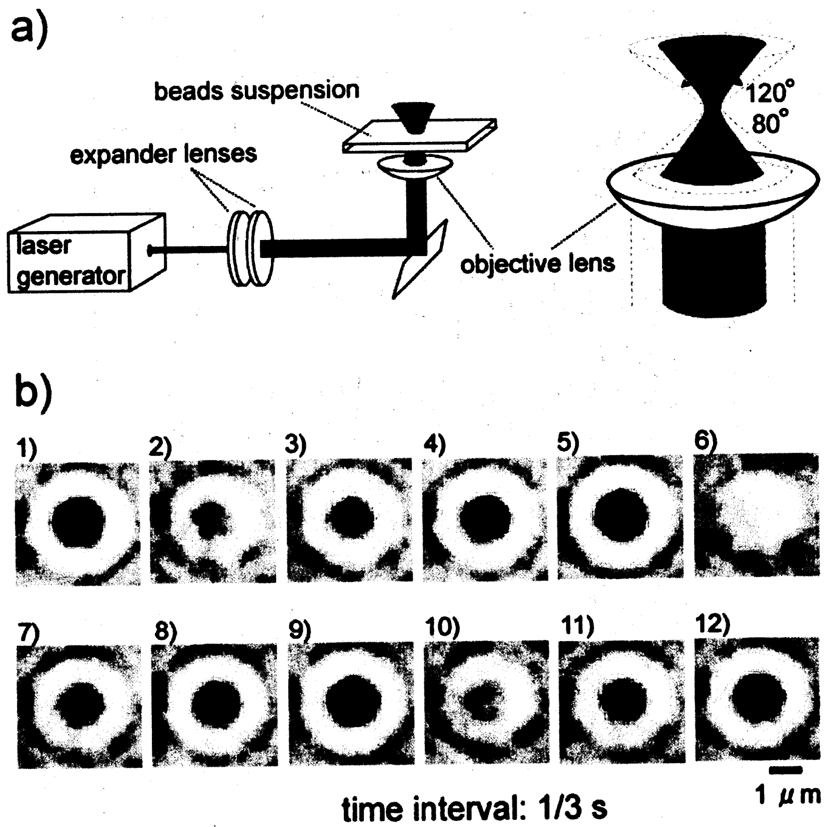

FIG 1. (a) Schematic representation of the experimental setup. The distance between the expander lenses was adjusted so as to decrease the focusing cone angle, *ca.* 80°. (b) Periodic growth and the burst of bead-cluster (black region), observed at 1.0 W of the laser power. Continues wave Nd:YAG laser (1064 nm, SL902T, Spectron) was used for the illumination. The mode of the laser is TEM_{00}, the profile of the laser has Gaussian distribution as confirmed by a beam profiler (Melles Griot). The laser beam was introduced into inverted microscope (TE-300, Nikon) through expander lenses and reflect mirrors, through 100x oil immersed objective lens. Suspension of negatively charged beads of polystyrene latex (0.20 μm in diameter) was purchased from Dow Chemical. The beads suspension, which contains 0.1 % solid, was situated between the glass plates. The thickness of the liquid was about 200 μm. Temperature was 20±2 °C.

charge. Without laser illumination, the beads disperse homogeneously in aqueous solution due to negative charge. Focused laser under standard angle 120° of the optical cone induces stational clustering of the beads.[11-13] By changing the cone angle from 120° to 80°, rhythmic change of growth and burst of the cluster is generated as in Fig. 1b. Where the dark circle corresponds to the dense clustering of the beads trapped on the focus.

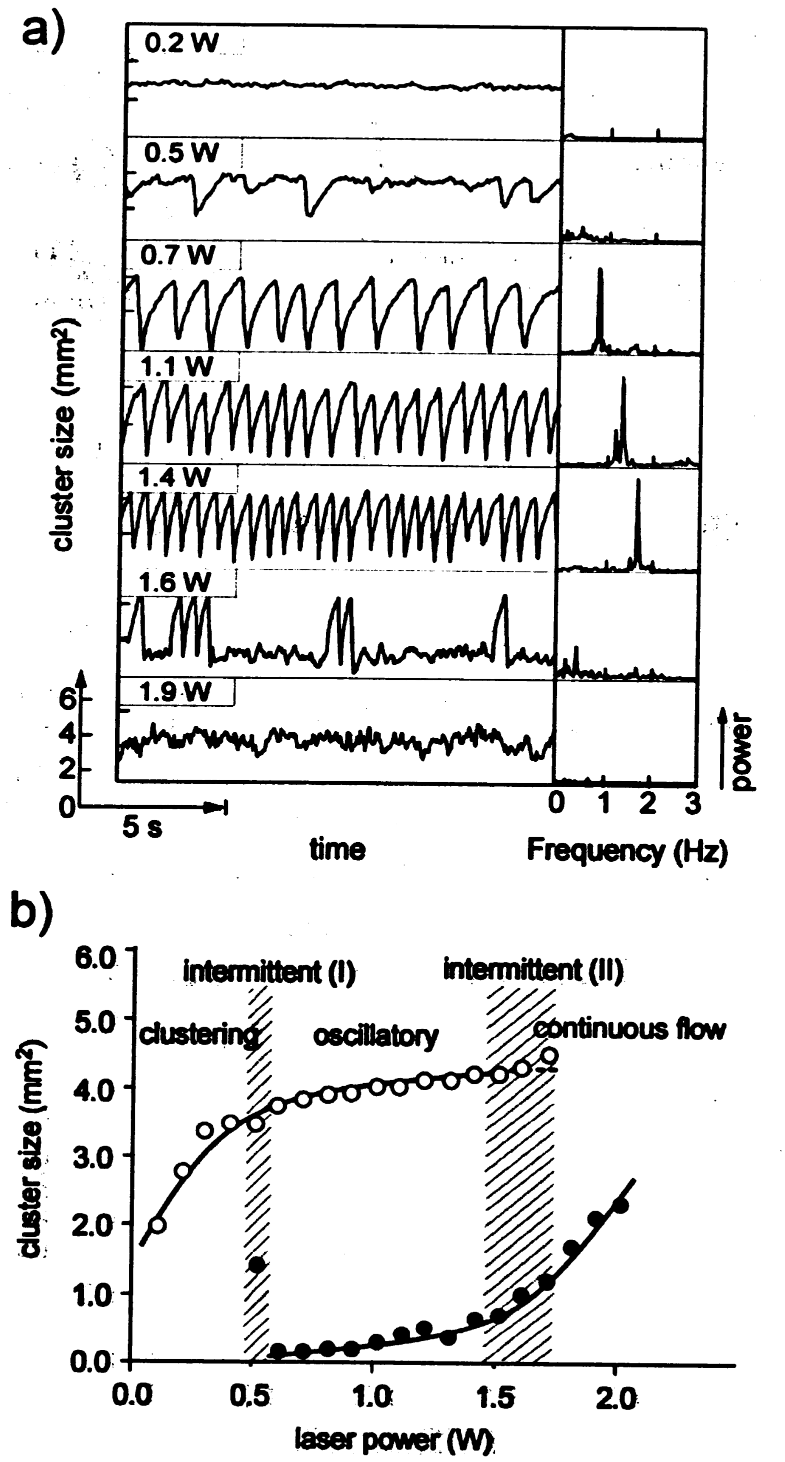

FIG 2. (a) Time-trace of the cluster area (left-handed side) with the time interval of 1/30 sec and the power spectrum by FFT analysis (right-handed side). (b) Diagram of clustering depending on the laser power. Closed circle and open circle indicate the maximum and minimum values in the oscillation on fluctuation of the cluster, respectively.

Figure 2a shows the time-trace of the cluster area (left-handed side), together with the frequency spectrum of the autocorrelation, or power spectrum of the Fourier Transformation (right-handed side). When the laser power is less than 0.4 W, small cluster isinduced and trapped in a stationary manner. With increase of the power, fluctuation of the cluster gradually grows (*e.g.*, at 0.5 W), Above 0.6W, rhythmic bursting is generated, where almost all of the beads are blown out at the occasion of the burst. Further increase of the power has the effect to increase the frequency of the bursting and the amplitude of the cluster area in a gradual manner. Around 1.6 W, the oscillation tends to be irregular. When the power is larger than 1.8 W, no oscillation is observed, *i.e.*, continuos flow of the beads-suspension is generated, without the formation of cluster on the focus. Fig. 2b shows the diagram of the state of beads-cluster as a function of the laser power.

The process of the growth and bursting of the beads cluster is schematically depicted in Fig. 3. (I) Driven by the radiation pressure, beads flow toward the focused region. (II) and (III), The cluster grows gradually. (IV) When the cluster grows up to a critical size, the cluster bursts, and the beads are flown away. Then, the growth of the cluster starts again. Such rhythmic change continues under stationary irradiation of laser. We have observed the side view of the cluster, by tilting the optical axis (data not shown). The above scheme on the rhythmic change has been actually confirmed from such observation of the time-dependent change of the morphology of the cluster.

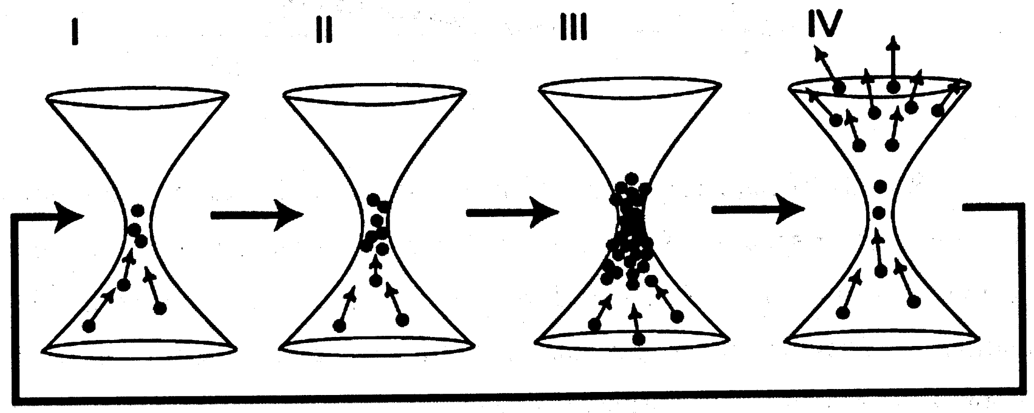

FIG 3. Schematic representation on the mechanism of periodic bursting.

Based on the above-mentioned experimental observation, we consider the following simple model. As for the growth process of the cluster, we adopt the kinetics as in eq. (2).

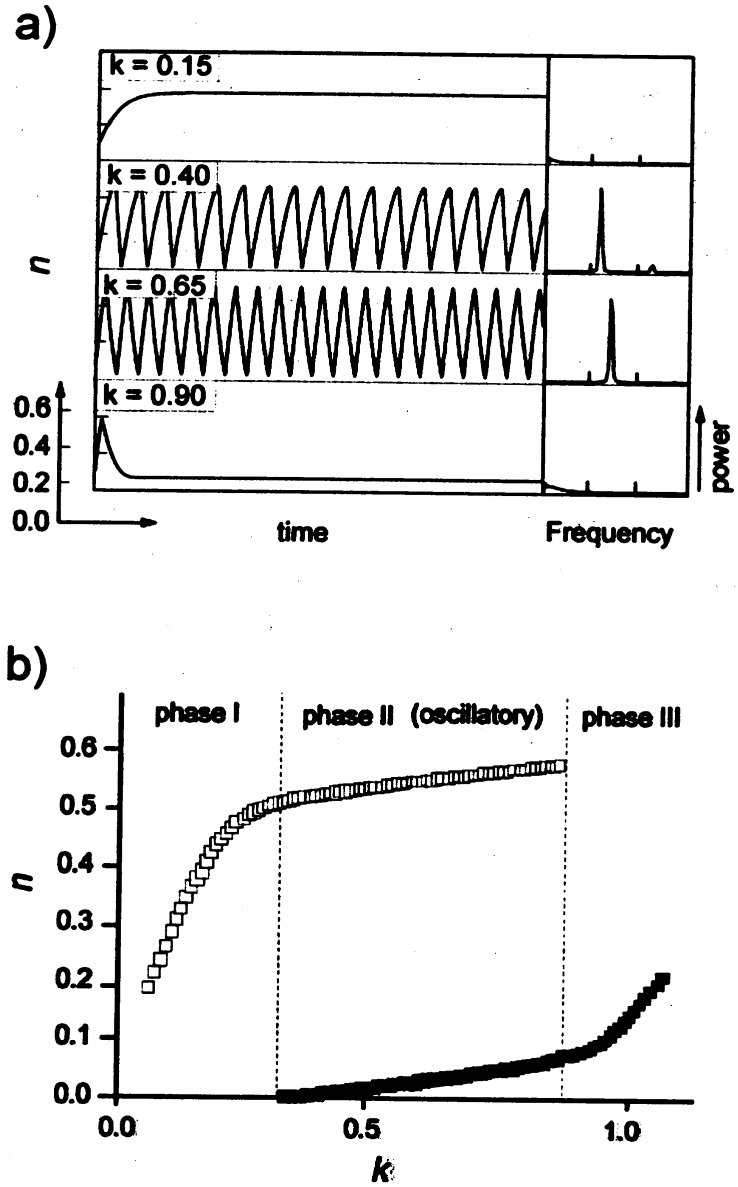

FIG 4. (a) Results of numerical simulation on the rhythmic bursting, time-trace of n (left) and the power spectrum (right). Forth Runge-Kutta method was used and the set of parameters, $a = 1.0$, $b = 1.0$, $c = 0.73$, $n_c = 3$, $\gamma = 0.5$, and $\eta = 3$, were chosen for the simulation. (b) Diagram of n depending on k. In oscillatory region (open square), the maximum and minimum were displayed in each k.

$$\dot{n} = k(n_c - n)^{\gamma} - u \tag{2}$$

e first term in eq. (2) represents the growth rate. *nc* is the upper critical number of beads i :luster, and k is a function of attractive force of the laser. As the cluster grows wit deformation from spherical symmetry, the actual growth rate is rather complicated. However, for simplicity we take a simple function to describe the growth rate by incorporating a single parameter γ to the equation. From the experimental observation, it is found that $\gamma \approx 0.5$. The second term, u, corresponds to the rate of escape from the cluster. As for the time dependant change of the flow rate, $\dot{u}$, we adopt the relationship as in eq. (3),

$$\dot{u} = n - (u - a)^{\eta} + cu - b, \qquad (3)$$

where a, b, and c are constants. In the growing phase the cluster size gradually increases. Accompanied with the growth, the effective light-pressure acting on the cluster increases. When the cluster grows to upper limit, the position of the cluster tends to be lifted. Owe to the narrowness of the potential near the focus, most of the beads become to be located out of the effective attractive potential (Fig. 3 III and IV). This causes a significant instability in the cluster. As a result, the cluster bursts, due to the intrinsic repulsive nature between the beads. Such a switching of the time-differential of escape rate $\dot{u}$ is represented in eq. (3), by taking $\eta = 3$ as the simplest choice of the parameter.

Figure 4a shows the time dependent change of the bead-number in a cluster and the power spectrum of FFT analysis, as is calculated from eqs. (2) and (3). In the simulation, we have changed k as a representative parameter of the laser power. The cluster stays stationary below a lower critical value of k. When k is between 0.35 and 0.89, the cluster begins to exhibit the periodicity of growth and bursting. It is noted that the amplitude increases and the periodicity decreases with the increase of k. Above upper critical value, 0.9, no oscillation of the cluster is shown. Figure 4b indicates the phase diagram depending on k. Such behavior of the clustering phenomenon in the simulation of Figs. 4a and 4b correspond well to the experimental trend, in spite of the simplification and roughness assumption in the modeling (eqs. (2) and (3)). It is expected by taking into account of adding noise and so on that the experimental result can be reproduced more correctly.

In conclusion, we showed the appearance of oscillatory instability in the cluster of beads under continuous laser illumination by choosing appropriate angle for the optical cone. The oscillation is caused owe to the cooperation between trapping force and scattering force exserted by the focused laser.

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